

PATENT
8007-1116

IN THE U.S. PATENT AND TRADEMARK OFFICE

In re application of

Takashi SUEYOSHI et al.

Conf. 1846

Application No. 10/594,221

Group 4145

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Examiner O. Ojurongbe

SILICON-CONTAINING CURING COMPOSITION AND HEAT CURED
PRODUCT THEREOF

DECLARATION UNDER RULE 132

Assistant Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Takashi SUEYOSHI, hereby declare as follows:

I am one of the inventors of the above-identified
U.S. patent application. My relevant background and
experience are set forth on the attached CV.

I have read the Official Action mailed November 7,
2008, and I am familiar with the present application. In
reviewing the Official Action, there does not appear to be any
appreciation for the both excellent heat resistance and
flexibility obtained with a silicon containing curing
comprising a silicon containing polymer with the recited Si-H
group only at terminal positions thereof or the recited Si-O-
Si bridge structure.

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To demonstrate the superior heat resistance and flexibility, I have conducted the following experiments which evaluate the structural differences between comparative examples and the present invention.

Comparative Example 3 is directed to a polymer containing a Si-H group in a repeating unit. In Synthesis Examples 17 and 18, the polymer of Comparative Example 3 is synthesized.

Comparative Example 4 corresponds to the adhesive composition of Koichiro as mentioned earlier, directed to polymer having no "Si-O-Si bridge structure". In Synthesis Example 19, the polymer of Comparative Example 4 is synthesized.

[Synthesis Example 17]

Ninety parts of dichlorodimethylsilane and 9 parts of dichloromethylsilane were mixed, and the mixture was delivered by drops into 100 parts of ion exchanged water. The reaction mixture was freed from the aqueous phase and polymerized at 250°C for 2 hours while removing the solvent by evaporation. To the resulting reaction solution were added 50 parts of dioxane and 5 parts of ion exchanged water, followed by stirring for 30 minutes. The precipitate thus formed was collected, and 50 parts of toluene was added to the precipitate. The solvent was removed by evaporation at 100°C

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under reduced pressure to give silicon containing polymer precursor 13. As a result of GPC analysis, the silicon containing polymer precursor 13 was found to have an Mw of 50,000.

[Synthesis Example 18]

Twenty-five parts of the silicon containing polymer precursor 13 obtained in Synthesis Example 17 and 2.5 parts of pyridine were added to a toluene solvent. To the mixture was further added 0.25 parts of phenyltrichlorosilane, followed by stirring at room temperature for 30 minutes and then at 70°C for 30 minutes. To the resulting mixture was added 2.5 parts of dimethylchlorosilane, followed by stirring at room temperature for 30 minutes and then at 70°C for 30 minutes and further followed by washing with ion exchanged water to remove pyridine hydrochloride to give silicon containing polymer 13B.

On the other hand, 25 parts of the silicon containing polymer precursor 3 obtained in Synthesis Example 3 and 2.5 parts of pyridine were added to a toluene solvent. To the mixture was further added 0.25 parts of phenyltrichlorosilane, followed by stirring at room temperature for 30 minutes and then at 70°C for 30 minutes. To the resulting mixture was added 2.5 parts of dimethylvinylchlorosilane, followed by stirring at room temperature for 30 minutes and then at 70°C for 30 minutes and

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further followed by washing with ion exchanged water to remove pyridine hydrochloride to give silicon containing polymer 13A.

The silicon containing polymers 13A and 13B were both found to have an Mw of 130,000 and, the proportion of a component with a weight average molecular weight of 1000 or less in these polymers was 0%.

[Comparative Example 3]

To 50 parts of an equal amount mixture of the silicon containing polymer 13A and the silicon containing polymer 13B was added 0.005 parts of a platinum-carbonylvinylmethyl complex as a curing reaction catalyst to prepare comparative silicon containing curing composition 3.

The comparative silicon containing curing composition 3 was cured at 250°C for 3 hours. The resulting cured product was evaluated for heat resistance in the same manner as in Examples of the present specification, and the 5% weight loss temperature was 375°C as shown in Table 1 below.

The tensile stress and elongation of the above comparative cured product were measured. The results of the measurement are shown in Table 1 below. In this Table 1 the measurement results of tensile stress and elongation of Examples 1-11 and Comparative Examples 1-2 of the present specification are also shown.

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As apparent from the Table 1 below, the comparative silicon containing curing composition 3 is excellent in heat resistance but inferior in flexibility, namely, both tensile stress and elongation are insufficient.

[Synthesis Example 19]

A hundred parts of dichlorodimethylsilane was delivered by drops into 100 parts of ion exchanged water. The reaction mixture was freed from the aqueous phase and polymerized at 250°C for 2 hours while removing the solvent by evaporation. To the resulting reaction solution were added 50 parts of dioxane and 5 parts of ion exchanged water, followed by stirring for 30 minutes. The precipitate thus formed was collected, and 50 parts of toluene was added to the precipitate. The solvent was removed by evaporation at 100°C under reduced pressure. To the resulting reaction solution was added 5 parts of pyridine, and the resulting mixture was divided into equal halves. To one of the halves was added 2.5 parts of dimethylchlorosilane, and 2.5 parts of dimethylvinylchlorosilane was added to the other, each followed by stirring at room temperature for 30 minutes and then at 70°C for 30 minutes and further followed by washing with ion exchanged water to remove pyridine hydrochloride to give silicon containing polymer 14B and silicon containing polymer 14A, respectively. As a result of GPC analysis, the

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silicon containing polymers 14A and 14B were both found to have an Mw of 30,000

[Comparative Example 4]

Fifty parts of the silicon containing polymer 14A, 50 parts of the silicon containing polymer 14B, and 1 part of 1,3,5,7-tetravinyl-1,3,5,7-tetramethylsiloxane were mixed, and to the mixture was further added 0.005 parts of a platinum-carbonylvinylmethyl complex as a curing reaction catalyst to give comparative silicon containing curing composition 4.

This comparative silicon containing curing composition 4 was cured at 250°C for 3 hours. The resulting cured product was evaluated for heat resistance in the same manner as in Examples of the present specification, and the 5% weight loss temperature was 282°C as shown in Table 1 below.

The above comparative cured product was evaluated for tensile stress and elongation. The results are shown in Table 1 below.

As apparent from the Table 1 below, the comparative silicon containing curing composition 4 has rather good flexibility but has inferior heat resistance.

TABLE 1

	Example										Comp. Example				
	1	2	3	4	5	6	7	8	9	10	11	1	2	3	4
Heat Resistance (5% weight loss temperature) (°C)	375	375	375	383	392	398	325	305	384	379	341	275	235	375	282
Tensile Stress (MPa)	9.0	7.0	3.0	10.0	12.0	13.0	8.0	7.0	6.0	7.0	8.0	3.0	1.0	3.0	3.0
Elongation (%)	100	100	160	90	80	70	110	130	140	150	160	180	200	50	150

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Takashi Sueyoshi
Takashi SUEYOSHI

Date March 4 2009